EFFECT OF SIMILAR ATOM SUBSTITUTION ON THE GLASS FORMING ABILITY OF Al_{86}Ni_{9}(Y, Sm)_{5} METALLIC GLASSES

VPLIV NADOMEŠČANJA MED SEBOJ PODOBNIH ATOMOV NA SPOSOBNOST TVORBE KOVINSKEGA STEKLA V ZLITINI Al_{86}Ni_{9}(Y, Sm)_{5}

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The effect of similar atom substitution on the glass forming ability (GFA) of Al_{86}Ni_{9}(Y, Sm)_{5} metallic glasses (MGs) was explored on the basis of the theory of the Fermi sphere-Brillouin zone interaction. Similar atom substitution (Sm) mainly affects the static structure between Al atoms and Y (Sm) atoms, changing the diameter of the pseudo-Brillouin zone (K_{p}). Its effect on the Fermi level and Brillouin zone size is characterized with spectroscopy experiments. The |\delta| = |K_{F} - 2K_{p}| criterion is used to evaluate the effect of the Sm element substitution on the GFA. This criterion can help us obtain the optimal GFA composition (Al_{86}Ni_{9}Y_{3.5}Sm_{1.5}) of Al_{86}Ni_{9}(Y, Sm)_{5} amorphous alloys, confirmed also by the experimental results.

Keywords: glass forming ability, Al-based metallic glasses, similar atomic substitution

1 INTRODUCTION

Al-based metallic glasses (MGs), with their excellent corrosion resistance and ultra-high specific strength, have been considered as significant potential engineering materials.1 However, because of their low GFA (glass forming ability), the largest size of Al-based bulk metallic glasses (MGs) currently reported is only 1.5 mm.2,3 The size limitation of Al-based MGs has significantly prevented their further applications.

Thermodynamic criteria, theoretical models, machine learning and high-throughput methods were proposed for solving this bottleneck problem of low GFA of MGs. First, it was understood that an enhancement in the GFA was closely related to decreasing the glass transition temperature (T_{g}).4,5 However, experimental results for the glass formation composition of Al-based MGs are often found to be in conflict with the currently available thermodynamic rules or criteria.6 Second, the GFA is fundamentally determined by the atomic structure. And both the chemical interaction and topological interaction between atoms codetermine the metallic glass stability.

Miracle et al. proposed an efficient cluster packing model,7,8 where GFA can be used to predict the GFA for MGs.9 An atomic electron tomography reconstruction method can be applied to experimentally determine the 3D atomic positions of amorphous alloys, providing direct experimental evidence to support the general framework of the efficient cluster packing model.10 But it is difficult to apply to the Al-based amorphous alloys since it does not consider the chemical interaction contribution to the MG stability. On this basis, an efficient atomic packing-chemistry coupled model was determined for the Al-TM-RE MGs system.11,12 And from the ab initio molecular dynamics and reverse Monte Carlo simulation, Sheng and Shi systematically explored the atomic packing in the Al-Ni-La, Al-Y-La and Al-Ni-Y-La amorphous alloys.13,14 Third, the machine learning and high-throughput approach are extremely powerful and efficient, having great potential for discovering new MGs with best GFA.15,16

Y. T. Sun et al. applied the support vector classification method to predict the GFA of binary amorphous alloys.17 X. Liu et al. investigated the glass formation through the data-driven machine learning technique and trained a backpropagation neural network model based
on a dataset covering thousands of ternary amorphous alloys. Applying a well-trained model, we can accurately identify the MG and non-MG classes and directly design compositions with best GFA. On the whole, the thermodynamic criteria, the above atomic structure models, computational simulation, machine learning and high-throughput methods can only reveal strong composition dependence of the GFA, but cannot predict the influence of similar atom substitution on the GFA for Al86Ni9(Y, Sm)5 MGs due to similar atomic sizes of the Y and Sm elements.

The role of electronic structure in the stability of MGs has long been recognized. Based on the Fermi sphere-Brillouin zone interaction, the Hume-Rothery stabilization mechanism was proposed by Nagel and Tauc. When the Fermi sphere with diameter \( K_{F} \) is in touch with the boundary of the pseudo-Brillouin zone with diameter \( K_{P} \) in the reciprocal space, \( 2K_{F} = K_{P} \), the phase stability is enhanced, coinciding with the minimum electronic density of the states at the Fermi level, \( E_{F} \). As for the Al-Ni-RE amorphous systems, \( 2K_{F} \) (the diameter of the Fermi sphere) is mainly affected by the sp-d electron hybridization between Al atoms and Ni atoms, which is similar to the compounds consisting of both transition metals and non-transition metals affected by hybridization. Furthermore, \( K_{P} \) (the diameter of the pseudo-Brillouin zone) is determined by the static atomic structure controlled primarily by the Al atoms and RE atoms. It may be possible to predict the GFA sensitivity to similar atom substitution for the Al86Ni9(Y, Sm)5 MGs by comparing the conditions of \( 2K_{F} = K_{P} \).

The purpose of this study is to investigate the effect of similar atom substitution on the GFA for the Al86Ni9(Y, Sm)5 MGs from the electronic structure aspect. The optimum glass-forming composition in ternary Al-based metallic glasses is Al86Ni9Y5. We consider the Sm element to change the Brillouin zone size and static atomic structure. The effects of the content of the Sm substitution on \( K_{F} \) is monitored by X-ray photoelectron spectroscopy (XPS), electron energy-loss spectroscopy (EELS) and X-ray diffraction (XRD) spectrometry. When \( 2K_{F} = K_{P} \), the phase stability is satisfied as much as possible for different contents of the Sm substitution, one may achieve a high GFA.

2 MODEL AND PROJECTED TREND FOR SIMILAR ATOM SUBSTITUTION IN THE Al86Ni9(Y, Sm)5 MGs

2.1 Fermi sphere-Brillouin zone interaction

The formation and stability of amorphous alloys were found to be influenced by the spherical-periodic resonance between the static atomic structure and the electronic system. The diameter of the Fermi sphere (\( 2K_{F} \)) can be obtained from a near-free-electron model in the electronic system (Equation 1):

\[
2K_{F} = 2\sqrt{3}\pi^{2}n_{0}Z
\]  

(1)

where \( n_{0} \) and \( Z \) are the mean valence and the mean atomic number density, respectively. For the Al-Ni-(Y, Sm) amorphous alloys, the electron hybridization effect between the Al-3p state and the Ni-3d state is extremely important, leading to a much lower density of conduction electrons that affects the Fermi sphere-Brillouin zone interaction. Consequently, the real electronic density of the states deviates from the nearly-free electron behavior from Equation (1) and it comes, instead, from the composite contribution of both the hybridization electrons between the Ni-3d state and Al-3p state (\( Z_{\text{hyb}} \)) and the nearly-free electrons (\( Z_{\text{FEM}} \)). \( 2K_{F} \) (the diameter of the Fermi sphere) can be given with Equation (2):

\[
2K_{F} = 2K_{F}^{\text{FEM}} - K_{F}^{\text{hyb}} = 2\sqrt{3}\pi^{2}n_{0}Z_{\text{FEM}} - 2\sqrt{3}\pi^{2}n_{0}Z_{\text{hyb}}
\]

(2)

Here, \( K_{F}^{\text{hyb}} \) and \( K_{F}^{\text{FEM}} \) come from the hybridization model and the free electron model, respectively. \( Z_{\text{hyb}} \) is the hybridized valence and \( Z_{\text{FEM}} \) is the mean valence in the free electron model.

The atomic number density is \( n_{0} = \rho N_{A} / M \) where \( N_{A} \) is the Avogadro’s number, \( \rho \) is the mass density of the MGs and \( M \) is the molar weight. In addition, the static atomic structure can be represented by the diameter of the spherical pseudo-Brillouin zone (\( K_{P} \)), while \( K_{F} \) is obtained from the position of the pseudo-Bragg peak in the structure factor (XRD):

\[
K_{F} = \frac{4\pi \sin \theta}{\lambda}
\]

(3)

Here, \( \theta \) is the location of the main peak in the XRD spectrum and the X-ray wavelength (\( \lambda \)) is 0.1542 nm.

The ideal resonance state is achieved at \( K_{P} = 2K_{F} \) in the reciprocal space, representing the equality of the two resonating subsystems. Consequently, the Fermi level is reached at the minimum electron density of the states.

2.2 Application to the Al86Ni9(Y, Sm)5 amorphous alloys

We considered the effect of the Sm element on the Fermi sphere-Brillouin zone interaction in the Al86Ni9Y5 ternary system. \( |\Delta| = |K_{P} - 2K_{F}| \) was used to identify the Sm element influence on the GFA. In this case, the hybridization between Al-3p and Ni-3d remains the same and \( K_{F} \) is unchanged, whereas \( K_{P} \) changes due to the static atomic structure of the Sm element substitution. When \( K_{P} \) is close to \( 2K_{F} \) (\( |\Delta| = 0 \)), the quasi-Brillouin zone touches the Fermi surface and the density of the states is minimized at the Fermi level. In this scenario, the GFA is influenced by the static atomic structure obtained due to the Sm element substitution.

Next, the effect of the Sm element substitution on \( K_{F} \) in the Al86Ni9Y5 ternary system was tracked during the
experiments. We obtained the $K_F$ value using Equation (2) where the key information was electron hybridization between the Al-3p state and Ni-3d state. $Z_{hp}$ and $Z_{dp}$ were evaluated using EELS and XPS. $K_F$ was obtained from the XRD spectrum according to Equation (3). Moreover, to assess the influence on the density of electron energy states at the Fermi level, we measured the electronic specific heat coefficient ($\gamma$), along with Sm microalloying. The electronic specific heat coefficient can be directly scaled with the density of states at the Fermi level.31

3 EXPERIMENTAL PART

$\text{Al}_{86}\text{Ni}_{9} (Y_{5-x}\text{Sm}_{x})_5$ ($x = (0, 1, 1.5, 2, 3, 4, 5)$) alloys were prepared by arc melting pure elements with nominal purities of above 99.9 % under a Ti-gettered argon atmosphere in a water-cooled copper crucible. The alloy ingots were melted six times to ensure compositional homogeneity. For rapidly solidified ribbons, samples with a cross-section of (0.03–0.05 × 3) mm were prepared using a single roller melt-spinning technique in an argon atmosphere. Wedge-shaped samples were prepared by casting molten alloys into a wedge-shaped mold with an included angle of 5°. The tip parts of the wedge-shaped samples were investigated by XRD using a Rigaku D/max 2400 diffractometer (Tokyo, Japan) with monochromatic Cu $K_\alpha$ radiation ($\lambda = 0.1542$ nm). We measured the low temperature specific heat $c_p$ using the Physical Property Measurement System (PPMS9000). XPS was performed using an ESCALAB250 photoelectron spectrometer and monochromatic Al $K_\alpha$ radiation ($h_\alpha = 1486.6$ eV). The surface of a sample was analyzed after short periods of Ar ion etching (4 keV) until the C 1s and O 1s signals were minimized. The size of ion beam was 2 mm, the sputter rate was 0.2 nm/min, and the time of ion etching was only a few seconds at room temperature. Thus, the above conditions of ion etching could not make the samples crystallize. Binding energies were calibrated using carbon contamination with a C 1s peak value of 284.6 eV. We used the Archimedes method to measure the density of melt-spin MGs. TEM samples were thinned by electropolishing in an electrolyte with an 80 % volume fraction of methanol and 20 % volume fraction of nitric acid at −35 °C. EELS was acquired using a field emission analytical TEM equipped with a Gatan Model 678 Imaging Filter (GIF), having a camera length of 80 mm and a spectrometer collection aperture of 3 mm. The ion milling removed the oxides from the surface and all spectra were examined for oxygen edges. We obtained normalized 3d white line intensity calculations from more than ten spectra to increase the accuracy.

4 RESULTS AND DISCUSSION

4.1 Effect of Sm on the GFA in $\text{Al}_{86}\text{Ni}_{9} (Y, \text{Sm})_5$ alloys

In order to monitor the electron hybridization between Al atoms and Ni atoms, we measured the chemical shifts of solute constituents in these alloys using XPS. As shown in Table 1 for $\text{Al}_{86}\text{Ni}_{9} (Y_{5-x}\text{Sm}_{x})_5$ ($x = (0, 1, 1.5, 2, 3, 4, 5)$) MGs, the Al 2p1/2 binding energies (BEs) in the alloys shifted to lower values by about 0.30 eV in comparison with pure Al, whereas the Ni 2p3/2 binding energies in the alloys moved to higher values by about 0.30 eV compared to those of pure Ni. Due to the high electrostatic energy, these shifts are unlikely to result from a real charge transfer, but, instead, can be interpreted as Al-3p-electrons "filling" the empty Ni-3d-states.32 In addition, we found no shift in the binding energy of Y 3d and Sm 3d, indicating there was no charge transfer between Al atoms and Y (Sm) atoms.

Table 1: Binding-energy difference between the pure element and $\text{Al}_{86}\text{Ni}_{9} (Y_{5-x}\text{Sm}_{x})_5$ alloys

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\Delta BE_{2p1/2} (\text{eV})$</th>
<th>$\Delta BE_{2p3/2} (\text{eV})$</th>
<th>$\Delta BE_{2p1/2} (\text{eV})$</th>
<th>$\Delta BE_{2p3/2} (\text{eV})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}Y_5$</td>
<td>0</td>
<td>−</td>
<td>−0.30</td>
<td>0.29</td>
</tr>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}Y_4\text{Sm}_1$</td>
<td>0</td>
<td>0</td>
<td>−0.29</td>
<td>0.30</td>
</tr>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}Y_4\text{Sm}_{1.5}$</td>
<td>0</td>
<td>0</td>
<td>−0.30</td>
<td>0.30</td>
</tr>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}Y_3\text{Sm}_2$</td>
<td>0</td>
<td>0</td>
<td>−0.30</td>
<td>0.29</td>
</tr>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}Y_2\text{Sm}_3$</td>
<td>0</td>
<td>0</td>
<td>−0.30</td>
<td>0.30</td>
</tr>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}Y\text{Sm}_4$</td>
<td>0</td>
<td>0</td>
<td>−0.29</td>
<td>0.30</td>
</tr>
<tr>
<td>$\text{Al}<em>{86}\text{Ni}</em>{9}\text{Sm}_5$</td>
<td>−</td>
<td>0</td>
<td>−0.29</td>
<td>0.29</td>
</tr>
</tbody>
</table>

In order to obtain some quantitative information on the charge transfer associated with hybridization, we further measured the white lines for L$_2$ and L$_1$ edges of the Ni element (Figure 1). For transition metals with unoccupied 3d states, the transition of an electron from a 2p state to 3d state leads to the formation of white lines. The atomic state changes from 2p$^3$d$^m$ to 2p$^3$d$(m+1)$ after the excitation of a 2p electron where $m$ represents the number of occupied 3d states. The L$_2$ lines correspond to the transition of 2p$_{1/2}$ to 3d$_{3/2}$3d$_{5/2}$ and the L$_3$ lines corre-
spond to the transition of 2p3/2 to 3d3/2 3d5/2. The intensities of L2 and L3 lines correlate with the unoccupied states in the 3d bands. Examining the white lines, we could obtain key information indicating that an electron transfer occurred between the outer d states of Ni atoms. It was concluded that the integrated intensities of white lines (the sum of L2 and L3 edge intensities) were normalized to the trailing background and they decreased nearly linearly with the increasing atomic number, which was reflected in the filling of the d states. Thus, the changes in the integrated white-line intensity (which is related to the trailing background) may be correlated directly with the changes in the number of d electron holes, i.e., a charge transfer.

We calculated the integrated white-line intensities for the L2 and L3 edges using the normalization method as shown in Figure 1. White lines were isolated by modeling the background with a double-step function. First, a straight line was fitted to the background, immediately following the L2 white line over a region of about 50 eV. Next, it was extrapolated into the threshold region. This line was further modified into a double step of the same slope with onsets located at the white-line maxima. We chose the ratio of the step heights as in accordance with the multiplicity of the initial states (four 2p3/2 electrons and two 2p1/2 electrons). Second, the white-line area above this step function was then divided by the area in a normalization window, 50 eV in width, beginning at 50 eV and passing the onset of the L3 white line. At last, applying the linear correlation of the normalized white-line intensity versus 3d occupancy, we could calculate the 3d electron occupancy of Ni in the Al86Ni9(Y5-xSmx)5 alloys, as follows:

$$I = 1.06(1 - 0.094n)$$

Here, n is the 3d occupancy of electrons per atom and I is the normalized white-line intensity. Equation (4) allows the calculation of n from the measured normalized white-line intensity. The 3d electron occupancy results are listed in Table 2. The obtained number of 3d electrons in Ni atoms indeed confirms the charge transfer from Al atoms to some outer 3d electrons of Ni atoms, which is in agreement with the chemical shifts from XPS.

Next, we calculated the $K_F$ of Al86Ni9(Y5-xSmx)5 (x = (0, 1, 1.5, 2, 3, 4, 5)) amorphous alloys. Taking $Z_{\text{Ni}} = 3$, $Z_{\text{Al}} = 2$, $Z_{\text{Ni}}$ in accordance with Equation (2). Using Equation (2), the $Z_{\text{FEM}}$ value of an alloy can be calculated with the contributions of all the constituent elements, i.e., $Z_{\text{FEM}} = \sum Z_i C_i$, where $Z_i$ and $C_i$ are the electron concentration of element i and the atomic fraction, respectively, and $Z_{\text{hyb}}$ can be obtained from $Z_{\text{hyb}} = C_{\text{Ni}} (n_{\text{Ni}} + n_{3\text{d}})$, in which $n_{\text{Ni}}$ and $n_{3\text{d}}$ represent pure Ni and the 3d occupancy electrons per Ni atom for Al86Ni9(Y, Sm)5 alloys, respectively.

Figure 2 shows x-ray diffraction scans for the Al86Ni9(Y5-xSmx)5 (x = (0, 1, 1.5, 2, 3, 4, 5)) wedge-shaped samples.

### Table 2: Electronic hybridization information from EELS for the Al86Ni9(Y5-xSmx)5 alloys

| Sample | $I_{\text{Ni}}$ | $n_{\text{Ni}}$ | $2K_{\text{F,hyb}}^{\text{EEM}}$ (1/nm) | $K_{\text{F,hyb}}^{\text{EEM}}$ (1/nm) | $K_F$ (1/nm) | $|\delta|$ (1/nm) | $D_C$ (μm) |
|--------|----------------|----------------|----------------------------------------|----------------------------------------|-------------|----------------|-----------|
| Al86Ni9Y5 | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 26.520 | 26.710 | 0.190 | 695 |
| Al86Ni9Y2Sm3 | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 26.520 | 26.650 | 0.130 | 903 |
| Al86Ni9Y2.5Sm2 | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 26.520 | 26.610 | 0.090 | 1000 |
| Al86Ni9Y2Sm | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 26.520 | 2.6390 | 0.130 | 899 |
| Al86Ni9Y4Sm1 | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 26.520 | 26.350 | 0.170 | 723 |
| Al86Ni9Y4Sm1.5 | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 26.520 | 26.300 | 0.220 | 659 |
| Al86Ni9Y3Sm2 | 0.2250 ± 0.0410 | 8.380 ± 0.041 | 34.320 | 7.800 | 3.6390 | 0.130 | 899 |

Figure 2: XRD patterns of the Al86Ni9(Y5-xSmx)5 wedge-shaped samples

Figure 3: Dependence of $K_F$ on the Sm atom addition in Al86Ni9Y5 alloys
shaped samples. The position of the main peak decreases with the increasing content of Sm. From XRD experiments we could evaluate the composition dependence of the $K_F$ value to assess the effect of the static structure. Using Equation (3), the $K_F$ values for the $\text{Al}_{86}\text{Ni}_9(\text{Y}_{5-x}\text{Sm}_x)_5$ amorphous alloys were calculated, as shown in Figure 3. The $K_F$ value decreases with the increase in the Sm addition and a $2K_F$ constant of 2.652 (black line). From Figure 4, $\text{Al}_{86}\text{Ni}_9\text{Y}_{3.5}\text{Sm}_{1.5}$ is expected to have the best GFA composition among the $\text{Al}_{86}\text{Ni}_9(\text{Y}_{5-x}\text{Sm}_x)_5$ alloys studied, as it displays the smallest $|\delta|$.

4.2 Experimental verification

Figure 2 presents the XRD tip parts of the wedge-shaped samples of the $\text{Al}_{86}\text{Ni}_9(\text{Y}_{5-x}\text{Sm}_x)_5$ ($x = (0, 1, 1.5, 2, 3, 4, 5)$) alloys where no observable crystalline peak is found in the X-ray pattern. Figure 4 shows the relationship between $|\delta|$ and critical thickness for glass formation ($D_c$) for different contents of Sm. The smaller the $|\delta|$ value, the higher is the GFA. This tendency of the GFA is in agreement with the previously predicted result. $\text{Al}_{86}\text{Ni}_9\text{Y}_{3.5}\text{Sm}_{1.5}$ is the best GFA composition, correlating to the minimum value of $|\delta|$. The correlation between $|\delta|$ and GFA for the $\text{Al}_{86}\text{Ni}_9(Y, \text{Sm})_5$ amorphous alloys clearly suggest that the $|\delta| = |2K_F - K_F|$ criterion is effective for predicting the Sm addition effect on the GFA.

The best $\text{Al}_{86}\text{Ni}_9(Y, Sm)_5$ ($x = (0, 1, 1.5, 2, 3, 4, 5)$) MG composition reported above indeed exhibits the lowest electronic density of states at the Fermi level. This conclusion was reached because we were able to assess the density of states at the Fermi level, $N(E_F)$, by measuring the electronic specific heat coefficient, $\gamma$. For comparison, the $\gamma$ values for the $\text{Al}_{86}\text{Ni}_9(Y, Sm)_5$ amorphous alloys are shown in Figure 5. Remarkably, the $\text{Al}_{86}\text{Ni}_9\text{Y}_{3.5}\text{Sm}_{1.5}$ MG with the best GFA is found to have the minimum $\gamma$ of all the alloys studied. This correlation between the $\gamma$ and the GFA indicates that the $|\delta| = |2K_F - K_F|$ criterion (i.e., the Fermi surfaces touching the quasi-Brillouin boundary) is indeed effective for controlling the stability of an amorphous structure and useful for predicting the effect of similar atom substitution (Sm) on the GFA in the $\text{Al}_{86}\text{Ni}_9(Y, Sm)_5$ MGs.

5 CONCLUSIONS

We give survey results on the effect of similar atom substitution on the GFA of the $\text{Al}_{86}\text{Ni}_9(Y, Sm)_5$ MGs based on the Fermi sphere-Brillouin zone interaction mechanism. For the Fermi sphere-Brillouin zone interaction in MGs, a Fermi sphere with diameter $2K_F$ is in touch with the pseudo-Brillouin zone boundary, $2K_F = K_F$, where the diameter of the Brillouin zone is defined by the principal diffraction peak located in the reciprocal space at $K_F$. The electronic density of states at the Fermi level is minimized so that the stability of the metallic glass is enhanced. The static structure changes of Al atoms and Y (Sm) atoms alter the diameter of the pseudo-Brillouin zone ($K_F$). The static structure changes of Al atoms and Y (Sm) atoms affecting the diameter of the pseudo-Brillouin zone are the reason for the similar atom substitution (Sm) effect on the GFA. The best GFA composition ($\text{Al}_{86}\text{Ni}_9\text{Y}_{3.5}\text{Sm}_{1.5}$) can be obtained when the condition $2K_F = K_F$ is achieved. The values of $K_F$ were confirmed by the experimental results of XRD, XPS, EELS and the electronic specific heat coefficient.

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